Water Quality Assessment and Monitoring Study: Literature Review of Chemical Tracers of Sewage

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King County

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Water Quality Assessment and Monitoring Study: Literature Review of Chemical Tracers of Sewage

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EXECUTIVE SUMMARY

This report documents a literature review conducted to better understand the state of the science regarding use of chemicals commonly present in sewage to trace the fate and transport of sewage in local surface waters. The review was done as part of King County's Water Quality Assessment and Monitoring Study, which was undertaken to explore ways to optimize water quality improvements in waterbodies where the County is planning combined sewer overflow control (CSO) projects.

Background

King County updates its CSO control plan about every five years. Before each update, the County reviews its entire CSO Control Program against conditions that have changed since the previous update. In September 2012, the King County Council passed Ordinance 17413 approving an amendment to King County's long-term CSO control plan. The amended plan includes nine projects to control the County's remaining 14 uncontrolled CSO locations in Lake Union/Ship Canal, Elliott Bay, and the Duwamish Estuary by 2030 to meet the Washington State standard of no more than one overflow per year on average. The recommended projects involve construction of underground storage tanks, green stormwater infrastructure, wet weather treatment facilities, or a combination of approaches.

Ordinance 17413 also calls for completion of a Water Quality Assessment and Monitoring Study (assessment) to inform the next CSO control plan update due to the Washington State Department of Ecology in 2019. The ordinance specified that the assessment answer the following questions:

1. What are the existing and projected water quality impairments in receiving waters (waterbodies) where King County CSOs discharge?
2. How do county CSOs contribute to the identified impairments?
3. How do other sources contribute to the identified impairments?
4. What activities are planned through 2030 that could affect water quality in the receiving waters?

Three additional questions will be addressed by the County's CSO planning team based partly on the results of the assessment:

5. How can CSO control projects and other planned or potential corrective actions be most effective in addressing the impairments?
6. How do various alternative sequences of CSO control projects integrated with other corrective actions compare in terms of cost, schedule, and effectiveness in addressing impairments?
7. What other possible actions, such as coordinating projects with the City of Seattle and altering the design of planned CSO control projects, could make CSO control projects more effective and/or help reduce the costs to WTD and the region of completing all CSO control projects by 2030?

Early in the assessment, a number of gaps were identified in the existing data that if filled, would provide critical information to answer study questions. Studies were identified to fill the data gaps and three studies were selected for implementation: sources and pathways of bacteria, contaminants of emerging concern, and chemical sewage tracers (the subject of this report).

Potential Uses of Chemical Sewage Tracers

Over the past decade or so, increasingly sophisticated analytical technology has enabled the analysis of a number of chemicals including hormones, pharmaceuticals, and personal care products. Researchers have been using these analytical methods (in conjunction with multivariate statistical tools) to determine if the sources of fecal contamination in surface waters are related to sewage or other sources.

Both inorganic and organic compounds specific to human sewage sources could potentially be used as tracers to identify the presence of human waste in receiving waters. A unique chemical (or group of chemicals) could be used to do the following:

- Detect untreated or inadequately treated sewage.
- Detect illicit connections of a sewage system into a stormwater system.
- Measure the effectiveness of infrastructure improvements after construction.
- Differentiate contributions of raw sewage in a particular CSO basin from the stormwater contribution to the basin.
- Monitor the fate and transport of sewage in surface waters.
- Estimate the relative contribution of raw sewage to any surface waters being monitored.

Study Approach and Results

The literature review looked at 14 primary and 27 secondary references representing studies in both the United States and other countries. The review focused on chemical tracers unique to human wastes, typically represented by artificial sweeteners, caffeine, nicotine, and certain pharmaceuticals and personal care products. Other types of tracers were also discussed in the literature (such as microbial source tracking, rhodamine dye, and colored dissolved organic matter). Although these types of tracers can serve as useful tools in addressing a specific study question, they were outside the scope of this review.

The review identified 29 compounds as potential sewage tracers. Of these compounds, caffeine, sucralose and acesulfame (artificial sweeteners), carbamazepine (anti-epilepsy drug), and metformin (diabetes drug) were viewed as the most promising:

- Caffeine has been used as a sewage tracer in many studies and could be an effective tracer if used in conjunction with other chemical tracers. It is easily detected using a common analytical method and its chemical characteristics in the environment may make it potentially useful to trace certain types of discharges.

- Although artificial sweeteners show promise for use as tracers, a cost-effective analytical method for these compounds is not available.
- Carbamazepine has been used by others as a sewage tracer because it is soluble and very stable in the aquatic environment, is not readily removed during the treatment process, and is specifically derived from human sources.
- Metformin also shows good potential as a tracer. It was frequently detected in surface waters at concentrations several times higher than detection limits during a King County survey of contaminants of emerging concern. However, the currently available analytical method is expensive.

To effectively use the compounds identified as potential sewage tracers as a means to address future management questions and concerns, specific study questions should be formulated. A full understanding of the scope, goals, and objectives for the use of chemical tracers is necessary to identify specific compounds for further evaluation.

An additional next step would be to identify cost-effective analytical methods for monitoring chemical tracers in the environment. Once identified, these methods could be used to characterize tracer concentrations and their contaminant pathways and to address questions about the fate and transport of sewage in the waterbodies of interest.

No single compound is suitable for addressing the variety of scientific inquiries for which tracers may be used. The use of several compounds has merit because multiple lines of evidence will improve confidence in the results. A group of compounds exhibiting a variety of chemical characteristics used under different environmental conditions would likely provide a broader range of information than a single tracer. King County will continue to monitor the state of sewage tracer research in the future.

Other Assessment Reports

This report is one of several reports that have been prepared as part of King County's Water Quality Assessment and Monitoring Study. Other reports are as follows:

- Reports describe existing conditions and long-term trends in three study areas—Lake Union/Ship Canal, Elliott Bay, and the Duwamish Estuary.
- A report documents the process used to assess identified data gaps for the study areas and select studies to fill prioritized gaps.
- Similar to this report, two reports discuss the methodology and results of selected new studies conducted to improve understanding of existing conditions: a study of bacteria in wet and dry weather and a survey of contaminants of emerging concern.
- A loadings report discusses present-day (2015) contributions of pollutants from various pathways, including stormwater runoff and CSOs, into the study areas and evaluates water quality impairments.

- A future loadings report assesses the potential of planned actions through 2030 such as CSO control projects and stormwater control and treatment projects to improve water quality.
- A final report summarizes these analyses and implications.

King County will use information from the Water Quality Assessment and Monitoring Study to inform the next CSO control plan update, including identification of opportunities to improve water quality outcomes, possibly reduce CSO control project costs, establish baseline conditions for post-construction monitoring of CSO control projects, and decide whether to pursue an integrated CSO control plan. The information from the assessment can also be used to inform regional efforts to continue to improve water and sediment quality.

ABBREVIATIONS AND ACRONYMS

CDOM	colored dissolved organic matter
CEC	contaminant of emerging concern
CRQL	contract required reporting limit
CSO	combined sewer overflow
Ecology	Washington State Department of Ecology
EPA	U.S. Environmental Protection Agency
LC-MS/MS	liquid chromatography-tandem mass spectrometry
MDL	method detection limit
MST	microbial source tracking
PMF	positive matrix factorization
PPCPs	pharmaceuticals and personal care products
PQL	practical quantitation limit
SAWPA	Santa Ana Watershed Project Authority
STRT	Scientific and Technical Review Team
USGS	U.S. Geological Survey
WTD	King County Wastewater Treatment Division

1.0 INTRODUCTION

This report documents a literature review conducted to better understand the latest scientific information available for use of common chemicals present in sewage to trace the fate of sewage and bacterial contamination in surface waters. It was prepared as part of King County's Water Quality Assessment and Monitoring Study, undertaken to explore ways to optimize water quality improvements in waterbodies where the County is planning combined sewer overflow control (CSO) projects.

The following sections describe the Water Quality Assessment and Monitoring Study, possible uses of sewage tracers, characteristics of effective tracers, and the scope the literature review.

1.1 Water Quality Assessment and Monitoring Study

King County owns and operates 38 CSO outfalls in the City of Seattle. The County's 2012 CSO control plan includes nine projects to control 14 uncontrolled CSOs by 2030 to meet the Washington State standard of no more than one overflow per year on a 20-year moving average. The recommended projects involve construction of underground storage tanks, green stormwater infrastructure, and/or wet weather treatment facilities. Four projects are in the Lake Union/Ship Canal area and five in the Duwamish Estuary and Elliott Bay areas.

Ordinance 17413, approving the CSO control plan, also calls for completion of a Water Quality Assessment and Monitoring Study (assessment) to inform the next plan update, which is due to regulators in 2019. In September 2013, the King County Council approved the assessment's scope of work through Motion 13966. The assessment includes a comprehensive scientific and technical analysis of water quality of the receiving waters ("study areas") where uncontrolled county CSOs discharge (Elliott Bay, Lake Union/Ship Canal, and the Duwamish Estuary). The study areas are shown in Figure 1-1.

A key component of the Water Quality Assessment and Monitoring Study was to complete water quality characterizations of the study areas using data previously collected from a variety of monitoring programs and studies. The characterizations included assessment of current water, sediment, and fish and shellfish tissue quality and other indicators of ecological health; evaluation of long-term trends in conditions over time; comparison to Washington State water and sediment quality standards to help identify impairments; and estimation of loadings to these waterbodies from contaminant pathways and expected future loadings following planned water quality improvement actions.

Early in the assessment, a number of gaps were identified in the existing data that if filled, would provide critical information on existing conditions in the study areas. Studies were identified to fill the data gaps and three studies were selected for implementation: sources and pathways of bacteria, chemical sewage tracers, and contaminants of emerging concern.

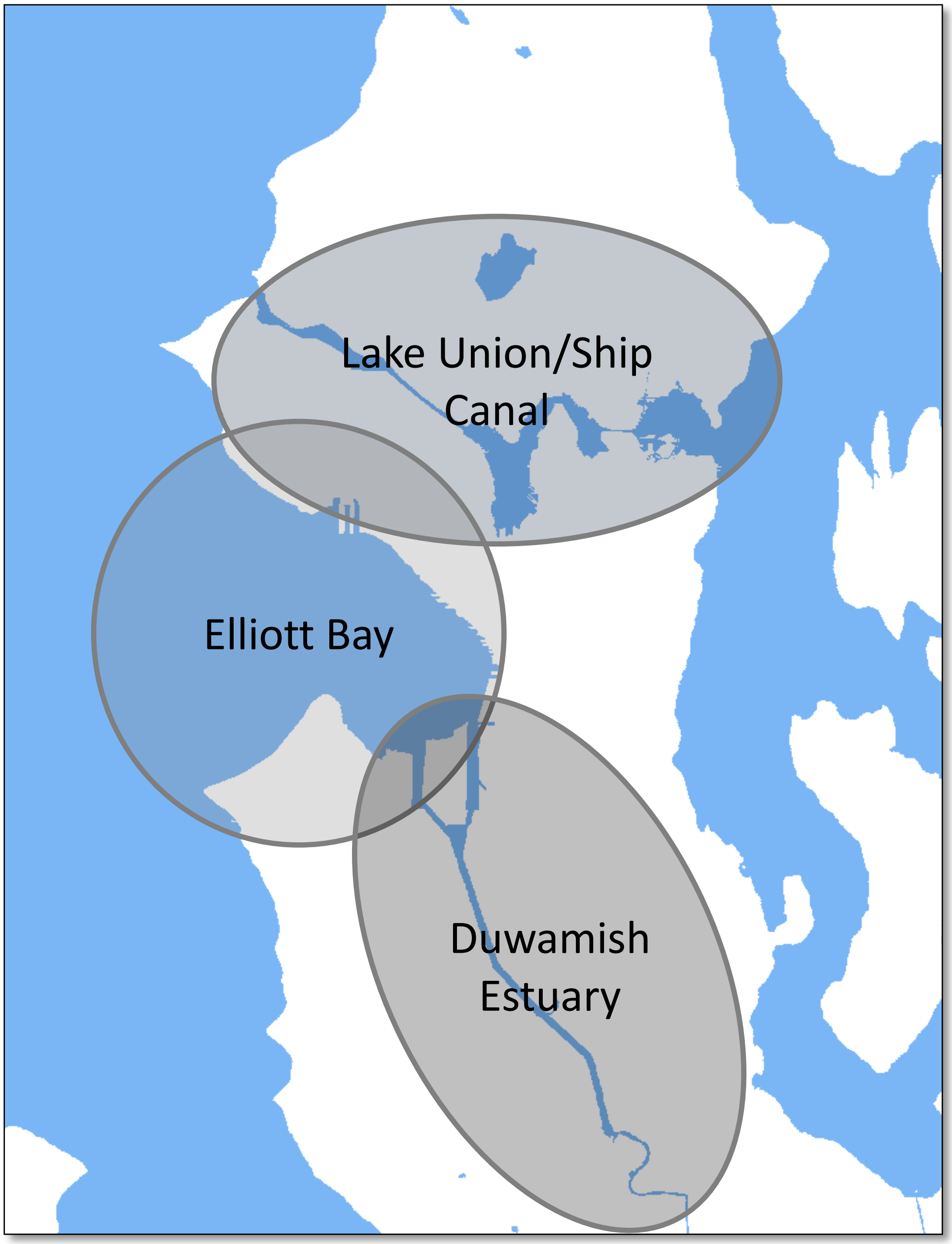

Figure 1-1. Lake Union/Ship Canal, Elliott Bay, and Duwamish Estuary study areas.

The Water Quality Assessment and Monitoring Study set out to generate information that will help answer the following study questions:

1. What are the existing and projected water quality impairments in receiving waters (waterbodies) where King County CSOs discharge?[1]

[1] "Impairments" is defined as water quality-related concerns.

2. How do county CSOs contribute to the identified impairments?
3. How do other sources contribute to the identified impairments?
4. What activities are planned through 2030 that could affect water quality in the receiving waters?
5. How can CSO control projects and other planned or potential corrective actions be most effective in addressing the impairments?
6. How do various alternative sequences of CSO control projects integrated with other corrective actions compare in terms of cost, schedule, and effectiveness in addressing impairments?
7. What other possible actions, such as coordinating projects with the City of Seattle and altering the design of planned CSO control projects, could make CSO control projects more effective and/or help reduce the costs to WTD (King County Wastewater Treatment Division) and the region of completing all CSO control projects by 2030?

The assessment addresses Questions 1 through 4. King County will use the information to inform the 2018 CSO control plan update, prioritize and sequence CSO control projects, establish baseline conditions for post-construction monitoring of CSO control projects, and decide whether to pursue an integrated plan based on U.S. Environmental Protection Agency (EPA) guidelines. Questions 5 through 7 will be addressed during the CSO control program update.

An external Scientific and Technical Review Team (STRT) was assembled to review methodology and results. Depending on assessment findings, the King County Council may decide to approve formation of an Executive's Advisory Panel of approximately 10 regional leaders. The panel would develop independent recommendations to the King County Executive on how planned county CSO control projects can best be sequenced and integrated with other projects to maximize water quality gains and minimize costs to ratepayers.

Table 1-1 shows elements of the assessment and their associated study questions, deliverables, and estimated timeframes. Figure 1-2 illustrates the flow of reports and how they will inform the CSO program review process. More information on the assessment is available at http://www.kingcounty.gov/environment/wastewater/CSO/WQstudy.aspx.

Table 1-1. Elements of the Water Quality Assessment and Monitoring Study.

Element	Applicable Study Question	Deliverable	Timeframe
Review and analyze existing scientific and technical data on impairments in Lake Union/Ship Canal, Duwamish Estuary, and Elliott Bay.	1	Area reports: • Elliott Bay • Lake Union/Ship Canal • Duwamish Estuary	2013–2017
Conduct targeted data gathering and monitoring to fill some of the identified gaps in scientific data on water quality in these receiving waters.	1,2,3	Data gaps analysis report[a] Data gap study reports: • Bacteria • Contaminants of emerging concern • Literature review of conservative sewage tracers	2014–2017
Identify and quantify the current (2015) pathways of contaminants into the receiving waters.	2,3	Loadings Report	2015–2017
Identify changes in contaminant loadings between 2015 and 2030, including the potential impact of planned corrective actions on identified impairments in the waterbodies.	1,2,3,4	Future Loadings Report	2015–2017
Summarize scientific and technical data collected and reviewed during the assessment.	1,2,3,4	Synthesis Report	2015–2017

[a] *Identification and Assessment of New Studies to Improve Understanding of Existing Conditions.*

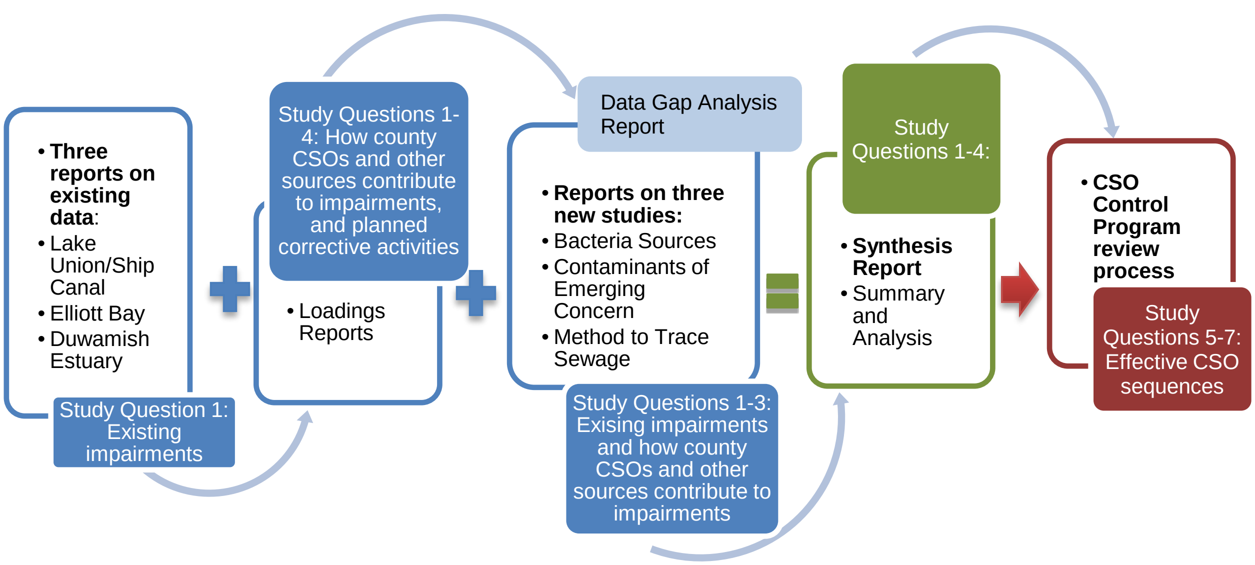

Figure 1-2. Reports and study questions answered as part of the Water Quality Assessment and Monitoring Study.

1.2 Possible Uses of Sewage Tracers

Over the past decade or so, increasingly sophisticated analytical technology has enabled the analysis of a number of chemicals including hormones, pharmaceuticals, and personal care products. Researchers have been using these analytical methods (in conjunction with multivariate statistical tools) to determine if the sources of fecal contamination in surface waters are related to sewage or other sources.

Both inorganic and organic compounds specific to human sewage sources could potentially be used as tracers to identify the presence of human waste in receiving waters. These compounds could be used to track and identify inputs of human waste from sources such as treatment plant effluent, CSOs, septic systems, and stormwater outfalls.

A unique chemical (or group of chemicals) could be used to do the following:

- Detect untreated or inadequately treated sewage.
- Detect illicit connections of a sewage system into a stormwater system.
- Measure the effectiveness of infrastructure improvements after construction.
- Differentiate contributions of raw sewage in a particular CSO basin from the stormwater contribution to the basin.
- Monitor the fate and transport of sewage in surface waters.
- Calculate the relative contribution of raw sewage to any surface waters being monitored.

Information from a tracer study may help address questions regarding contamination pathways, such as when treated and untreated CSOs are near each other, and help assess the efficiency of the County's wet weather treatment facilities. Tracer information may also help identify sewage inputs into a stormwater system and where these inputs may originate. Pharmaceutical tracers may also help local health officials better understand illicit drug use patterns, which are difficult to study.

1.3 Characteristics of Effective Sewage Tracers

Requirements of an effective human waste tracer vary widely depending on the study questions and the environmental conditions. Several studies (Gasser et al., 2010; Kasprzyk-Hordern et al., 2009; and Nakada et al., 2008) have identified specific characteristics necessary for a chemical to be effective as a sewage tracer for surface water, groundwater, or estuarine environments. According to the literature, compounds identified as a sewage tracer should possess the following characteristics:

- Abundant in sewage and only present in human waste streams.
- Not present in waters without human waste discharges.
- Detectable at levels well above method detection limits (MDLs). Concentrations near the MDL are inherently more uncertain than higher concentrations, which are closer to the midpoint of the instrument calibration range.

- Does or does not undergo significant biological or photolytic degradation. Both types of tracers can be useful.
- High water solubility, low partitioning coefficient (K_{ow}), and low volatility to minimize tracer losses to sedimentation, and the atmosphere.[2]
- Any seasonal changes in human use and excretion patterns are known and predictable.
- Depending on the specific study question, the removal efficiency or lack of removal through primary, secondary, or tertiary treatment processes is well understood.

1.4 Scope of the Review

This literature review was conducted to better understand if a chemical tracer or tracers could provide information on the fate of sewage in surface waters and to investigate the possibility of testing for a chemical tracer routinely as part of the King County monitoring program, thereby tracking tracers over time and in a variety of environmental conditions.

The identification of useful human sewage tracers is complicated by a number of variables such as sporadic or unpredictable use of some compounds, varying degradation rates, and uncertainty of the origins of the compounds. For example, untreated stormwater or illicit vessel sewage discharges may occur in the vicinity of CSOs making it difficult to connect a specific source to a detected compound.

To alleviate some of these uncertainties, a suite of tracers with known characteristics were identified during the project scoping process. The review focused on chemical tracers unique to human wastes, typically represented by artificial sweeteners, caffeine, nicotine, and certain pharmaceuticals and personal care products (PPCPs) (Buerge et al., 2008; Van Stempvoort, 2011). Other types of tracers are discussed in the literature, such as microbial source tracking (MST), rhodamine dye, and colored dissolved organic matter (CDOM). Although these types of tracers can serve as useful tools in addressing a specific study question, they were outside the scope of this review.

[2] A low K_{ow} means that the chemical is less likely to adsorb to sediment or tissue and stay in solution in the water column

2.0 SUMMARY OF STUDIES REVIEWED

This chapter summarizes the sewage tracer studies that were reviewed. The review included 14 primary references and 37 secondary references. It was not intended to be exhaustive, but rather an overview of relevant research on the subject of sewage tracers and their use. The summaries are organized by area: Pacific Northwest, other parts of the United States and in Canada, and Switzerland and Japan.

2.1 Pacific Northwest

The following studies conducted in the Pacific Northwest were reviewed:

- Johnson et al. (2004) conducted a screening analysis of PPCPs in wastewater treatment plant effluents, groundwater wells, and creeks in Northwestern Washington. Of the 24 compounds analyzed, 16 were consistently detected in effluents and only caffeine, nicotine, and metformin were detected in samples from nearby creeks and groundwater wells. The authors concluded that additional testing of these three chemicals for their use as potential sewage tracers was needed.
- In August 2008, the Washington State Department of Ecology (Ecology) and EPA conducted a one-day screening survey to characterize PPCPs at five municipal wastewater treatment plants in the Pacific Northwest (Lubliner et al., 2010). Wastewater influent, secondary effluent, tertiary effluent, and biosolids were analyzed to assess removal efficiencies of 172 organic compounds including PPCPs, hormones, steroids, and semivolatile organic compounds. EPA-approved analytical methods with low detection limits in the 2−10 ng/L Practical Quantitation Limit (PQL) range were used to measure PPCP concentrations. PPCPs were detected in all samples. Carbamazepine (anti-epileptic), fluoxetine (selective serotonin reuptake inhibitor – antidepressant), and thiabendazole (for treatment of intestinal nematodes – de-wormer) were relatively unaffected by the treatment technologies in use. While not the study focus, the authors concluded that these three PPCPs could potentially serve as sewage tracers in local waterbodies.
- In 2012, Johnson et al. conducted a study of PPCPs, hormones, and sterols in reclaimed water and groundwater in Washington state. Samples were collected from three locations including the reclaimed water facilities at Lacey, Quincy, and Yelm. These facilities use reclaimed water to recharge local groundwater supplies. Of the 145 compounds analyzed, 73 were detected in reclaimed water and only 15 were detected in groundwater samples. The compounds most frequently detected were drugs (or their metabolites) used to treat high blood pressure, followed by antidepressants and antibiotics. The authors concluded that carbamazepine, meprobamate (anti-anxiety drug), and sulfamethoxazole (antibiotic) would be useful as groundwater tracers at these facilities. These three compounds were detected in all reclaimed water and groundwater samples collected in the recharge area at each facility.
- Investigators at the University of Washington Tacoma Center for Urban Waters collaborated with the Kitsap Public Health District to evaluate the utility of using a

suite of contaminants of emerging concern (CECs) as tracers of bacterial contamination near the Puget Sound shoreline (James et. al., 2014). Samples from small freshwater discharges were collected at approximately 20 sites and analyzed for a suite of over 20 CECs commonly associated with human sewage, a suite of pesticides, and a number of other compounds of interest. The authors examined sucralose (an artificial sweetener) as one of several possible tracer compounds. Sucralose has shown good potential for tracing failed septic systems.

- King County conducted a preliminary presence-absence survey of a number of pharmaceuticals in ambient waters in the Lake Union/Ship Canal, Elliott Bay, and the Duwamish Estuary (King County, 2017) as part of the Water Quality Assessment and Monitoring Study. Metformin was detected and shows good potential for being used as a tracer because of its concentrations in surface waters. Concentrations as high as 786 ng/L were detected, which is well above the 3 ng/L detection limit. The frequency of detection was 52.9 percent, which is greater than most of the other compounds in the study.

2.2 Other Parts of the United States and in Canada

The following studies that examined potential sewage tracers in other parts of the United States or in Canada were reviewed:

- Kolpin et al. (2002) conducted the first comprehensive national surface water reconnaissance testing of pharmaceuticals, hormones, and other organic wastewater contaminants in the United States. This U. S. Geological Survey (USGS) study analyzed 95 compounds in water samples from 139 streams between 1999 and 2000. Of the 95 analytes tested, 82 were detected. The most frequently detected compounds were coprostanol (fecal sterol), cholesterol (sterol), N, N-diethyltoluamide (DEET insect repellant), caffeine (stimulant), triclosan (disinfectant), tri(2-chloroethyl)phosphate (flame retardant/plasticizer), and 4-nonylphenol (surfactant used in detergents – antioxidant). The study documented that detectable levels of the contaminants occur in streams throughout the United States. Many of these compounds passed through wastewater treatment processes and only a few were associated with stormwater or surface runoff.
- In 2005, EPA and USGS scientists conducted a national study to determine potential indicators of human waste (Glassmeyer et al., 2005). This study collected and analyzed water quality samples from upstream and downstream of 10 wastewater treatment plants located across the United States. One effluent sample was also collected from each treatment plant. Samples were analyzed for 110 organic chemicals to determine if a correlation existed between the presence of these chemicals and known human waste sources. The number of detected compounds at the sampling sites ranged from 3 at a background location to 50 in an effluent sample. Results from this study indicate that over 35 different chemicals may be useful as indicators of human wastewater sources. Carbamazepine, diphenhydramine (antihistamine), caffeine, and coprostanol were particularly noteworthy as potential tracers of human sewage contributions.

- Other USGS researchers, Benotti and Brownawell (2007), investigated a wide range of PPCPs in Jamaica Bay, an estuary in New York City and Nassau County, New York. This estuary is heavily influenced by wastewater effluent discharges (multiple treatment plants, large CSOs, and one large landfill). The main objective of this study was to demonstrate the usefulness of several persistent compounds as sewage tracers. The study also assessed degradable compounds, such as caffeine, as indicators of CSO (or untreated sewage) inputs into a coastal system. Caffeine, cotinine (nicotine metabolite), nicotine, and paraxanthine (caffeine metabolite) were detected most often. Carbamazepine and sulfamethoxazole (antibiotic) were also frequently detected and showed little evidence of removal during the treatment process. Because carbamazepine and sulfamethoxazole exhibited very low wastewater treatment removal rates, researchers concluded these pharmaceuticals would be useful as tracers of treated sewage effluents.
- In 2012, Canadian researchers evaluated PPCPs and caffeine as possible indicators of sewage contamination in drinking water sources (Daneshvar et al., 2012). The results from this study showed carbamazepine had the highest frequency of detection (> 99 percent) followed by caffeine, naproxen (anti-inflammatory/pain reliever), trimethoprim (antibiotic), and gemfibrozil (lipid regulator). The authors also measured a significant difference in the removal efficiency between carbamazepine and caffeine. Caffeine was more readily removed during the treatment process. The researchers concluded that caffeine would be useful as a tracer for untreated sewage contamination and that carbamazepine may be useful as a tracer for treated wastewater.
- California researchers Eaton and Haghani (2012) used MWH Laboratories' direct online extraction/analysis methodology that includes LC-MS/MS (liquid chromatography-tandem mass spectrometry) instrumentation to assess approximately 90 analytes of interest from lists of recommended indicators of sewage. The sampling at 17 locations was part of the Santa Ana Watershed Project Authority (SAWPA) effluent monitoring program in summer 2010 and 2011. Both receiving waters of the Santa Ana River and effluents discharged from wastewater treatment plants were sampled. Recommended compounds were also compared to studies across the country from various source types (such as drinking water and distribution systems) to determine the best indicators. Researchers concluded that none of the chemicals on the lists from SAWPA, the California State Water Resources Control Board, the Water Research Foundation, and the National Water Research Institute were suitable and that analyzing a larger list of potential sewage tracers was necessary to reduce uncertainty.

2.3 Switzerland and Japan

The following studies conducted in Switzerland and Japan were reviewed:

- In 2003, Buerge et al. conducted an in-depth study to evaluate the potential for caffeine to serve as a sewage tracer in surface waters. This study evaluated caffeine concentrations in wastewater treatment plant influents and effluents, receiving waters, pristine mountain lakes, and moderately polluted lakes and rivers in the mid-land region in Switzerland. The authors concluded that caffeine is an anthropogenic chemical used only by humans and therefore is a good indicator of wastewater inputs. Further research concluded that caffeine can be used more specifically as an indicator of untreated wastewater discharged as CSOs (Buerge et al., 2006).

- In 2008, Buerge et al. conducted a study of nicotine and its metabolites as potential tracers to detect the presence of domestic wastewater in Switzerland. The authors found that concentrations of nicotine metabolites correlated well with known wastewater inputs into lakes and concluded that three of these metabolites—cotinine, 3′-hydroxycotinine, and N-formylnornicotine—would serve as good wastewater tracers.
- In 2009, Buerge et al. studied four artificial sweeteners (acesulfame, cyclamate, saccharin, and sucralose) to examine their occurrence and fate in wastewater, surface waters, groundwater, and drinking water in Switzerland. The researchers also evaluated these sweeteners as potential chemical markers of domestic wastewater in groundwater and concluded that acesulfame met their desired criteria for such use.
- Researchers in Japan have also evaluated various contaminants as potential sewage tracers in different environments. For example, Nakada et al. (2008) examined 13 PPCPs for their efficacy as water-soluble molecular markers of sewage in rivers, groundwater, and coastal environments. The researchers were particularly interested in a potential tracer's stability in estuarine environments. They studied local groundwater, spring water, 37 major rivers, and the Tamagawa Estuary near Tokyo. Results indicate that crotamiton (anti-itch medication for the treatment of skin mites) and carbamazepine would be effective tracers in freshwater and coastal marine environments. The authors also recommended combining these tracers with more unstable PPCPs such as caffeine to help detect inputs from poorly treated or untreated sewage.

3.0 COMMONLY CITED COMPOUNDS

The practice of tracing human waste streams has been well documented by studies that identify several chemical tracers that are relatively stable and mobile in both surface and groundwater environments. The studies cited in this literature review document the presence of potential tracers detected in surface water, groundwater, or marine environments. Most of these efforts focused on caffeine, artificial sweeteners, fecal sterols, and PPCPs. These are typically the most prevalent and/or most frequently detected compounds in a variety of environmental systems. Table 3-1 lists the chemicals that were most frequently cited in the literature as suitable tracers.

Table 3-1 Most frequently cited compounds in the literature as potential tracers of some types of human wastewater or sewage.

Chemical Name	Classification/Use	Reference
Acesulfame	Artificial sweetener	Buerge et al., 2009
Acetaminophen	Antipyretic	Glassmeyer et al., 2005; Doyle et al., 2009
Caffeine	Stimulant	Glassmeyer et al., 2005; Hyer, 2007; Nakada et al., 2008; Benotti and Brownawell, 2007; Buerge et al., 2003; Daneshvar et al., 2012; Seiler et al.,1999; Kolpin et al., 2002.; Doyle et al,. 2009.
Carbamazepine	Anti-epileptic	Glassmeyer et al., 2005; Nakada et al., 2008; Lubliner et al., 2010; Benotti and Brownawell, 2007; Daneshvar et al., 2012; Clara et. al., 2004; Doyle et al., 2009; Johnson et al., 2012.
Cholesterol	Sterol	Glassmeyer et al., 2005; Kolpin et al., 2002.
Codeine	Analgesic	Glassmeyer et al., 2005.
Cotinine	Nicotine metabolite	Glassmeyer et al., 2005; Benotti and Brownawell, 2007; Buerge et al., 2008; Doyle et al., 2009; Hyer, 2007.
Coprostanol	Fecal sterol	Glassmeyer et al., 2005; Kolpin et al., 2002.
Crotamiton	Antipruritic	Nakada et al., 2008.
Diphenhydramine	Antihistamine	Glassmeyer et al., 2005
Ethenzamide	Anti-inflammatory	Nakada et al., 2008.
Fenoprofen	Anti-inflammatory	Nakada et al., 2008.
Fluoxetine	Serotonin reuptake inhibitor	Lubliner et al., 2010
Gemfibrozil	Blood lipid and cholesterol altering	Daneshvar et al., 2012.
Ibuprofen	Anti-inflammatory	Doyle et al., 2009; Nakada et al., 2008.
Ketoprofem	Anti-inflammatory	Nakada et al., 2008.
Mefenamic acid	Anti-inflammatory	Nakada et al., 2008.
Meprobamate	Anti-anxiety	Nakada et al., 2008; Johnson et al., 2012.

Chemical Name	Classification/Use	Reference
Metformin	Diabetes drugs	Blair et al., 2013
N-N-diethyltoluamide (DEET)	Insect repellant	Glassmeyer et al., 2005; Kolpin et al., 2002; Nakada et al., 2008.
Naproxen	Anti-inflammatory	Nakada et al., 2008; Daneshvar et al., 2012.
Nicotine	Tobacco ingredient	Benotti and Brownawell, 2007; Buerge et al., 2008.
Paraxanthine	Caffeine metabolite	Nakada et al., 2008.
Propyphenazone	Analgestic and antipyretic	Nakada et al., 2008.
Saccharin	Artificial sweetener	Buerge et al., 2009; Van Stempvoort, 2011; Mawhinney et al., 2011.
Sucralose	Artificial sweetener	Buerge et al., 2009; Van Stempvoort, 2011; Mawhinney et al., 2011; Oppenheimer et al., 2011.
Sulfamethoxazole	Antibiotic	Glassmeyer et al., 2005; Kolpin et al., 2002; Barnes et al., 2008; Benotti and Brownawell, 2007; Johnson et al., 2012.
Triclosan	Disinfectant	Glassmeyer et al., 2005; Nakada et al., 2008; Kolpin et al., 2002; Hyer, 2007.
Trimethoprim	Antibiotic	Glassmeyer et al., 2005; Daneshvar et al., 2012.

4.0 DISCUSSION

This chapter discusses in more detail individual chemicals or groups of chemicals that consistently emerged as potential sewage tracers in the literature: caffeine, artificial sweeteners, and PPCPs.

4.1 Caffeine

Caffeine has been used as a tracer of human waste for many years (Buerge et al., 2003; Seiler et al., 1999). Two studies detected caffeine in over 70 percent of samples collected downstream of wastewater treatment plants (Kolpin et al., 2002; Glassmeyer et al., 2005).

Although caffeine had been shown to be a suitable marker for sewage contamination in surface water, it does present some challenges. For example, once in the environment, caffeine becomes unstable, degrades rapidly (Nakada et al., 2008), and is broken down rapidly during the wastewater treatment process (Benotti and Brownawell, 2007; Burge et al., 2003). Caffeine concentrations as high as 300 µg/L have been reported in raw sewage and as low as 0.1 µg/L in treated wastewater effluent (Burge et al., 2003).

Detection of caffeine in waterbodies that do not receive effluent inputs limits its utility as a sewage tracer. Despite its high removal rate (> 99 percent) during treatment, much lower concentrations of caffeine are still detected in rivers, lakes, urban streams, and estuaries ranging from 3 ng/L to 1,500 ng/L (Buerge et al., 2003; Doyle et al., 2009). Therefore, caffeine should be considered only for the detection of untreated wastewater contamination (Daneshvar et al., 2012).

Caffeine is potentially useful as a tracer for untreated discharges in surface waters that also receive treated discharges. Because it is easily removed by the treatment process, it can be used to help differentiate between treated and untreated discharges. It quickly breaks down in the environment, making it possible to help differentiate between older and more recent discharges. Rapid degradation, however, poses challenges for completing field sampling and analysis in time. In addition, many sources and pathways exist and can confound tracing this compound back to individual sources.

4.2 Artificial Sweeteners

Artificial sweeteners (sucralose and acesulfame) show promise as tracers. Researchers have found that these chemicals are abundant in sewage, although geographical preferences may play a role in the distribution of individual sweeteners. Sweeteners are not removed during the treatment process and are persistent in the environment. Analytical methods are in the research phase. Their primary sources are food additives and therefore their key pathway to the environment is human waste.

Sucralose (Splenda® in the United States) is used in trace amounts because it is 600 times sweeter than sucrose. It passes through the human digestive system nearly unchanged. Of the ingested dose, 95–98 percent is excreted intact (Buerge et al., 2009; Loos et al., 2009). Sucralose has been used as a human waste tracer in surface waters across Western Europe (Buerge et al., 2009) and Canada (Van Stempvoort et al., 2011). It has been detected in drinking water supplies across the United States (Mawhinney et al., 2011).

Acesulfame is also a widely used artificial sweetener; 100 percent of this compound is excreted from the human digestive system (Buerge et al., 2009). Acesulfame has been used as both a surface water and groundwater tracer in many countries (Buerge et al., 2009; Mawhinney et al., 2011; Van Stempvoort et al., 2011). It has been detected in groundwater in Zurich, Switzerland, where aquifers are recharged with river water containing treated sewage from treatment plant effluent (Buerge et al., 2009).

A variety of other artificial sweeteners have been used as tracers, but issues with degradation and detection frequency in treatment plant effluents suggest they may be applicable only to particular study questions. For example, saccharin is used worldwide in many beverages and personal care products; 90–100 percent of the compound is excreted after ingestion. Saccharin is used in many products produced in the United States, but not as frequently as sucralose and acesulfame.

Buerge et al. (2009) found that acesulfame and sucralose concentrations were not significantly altered by the wastewater treatment process while removal rates for saccharin and cyclamate were 90 and 99 percent, respectively, in treatment facilities using an activated sludge process. Thus, the ratios of these different compounds could aid in understanding whether the sources to a particular waterbody are from treated or untreated effluents and their relative contributions.

In addition, compounds could be washed off in stormwater from impervious surfaces (streets and sidewalks) where beverages containing these sweeteners have been spilled. Quantifying spills can be difficult, and additional loading estimates would need to be determined in order to properly characterize this pathway. Once a reliable analytical method is available, further characterization of surface waters and pathways of pollutants to surface waters could be done.

4.3 Pharmaceuticals and Personal Care Products

The occurrence of PPCPs in ambient surface waters has been reported nationwide. PPCPs have been extensively studied by numerous investigators (Glassmeyer et al., 2005; Johnson et al., 2012; Nakada et al. 2008); however, very few have been identified as a viable sewage tracer.

Two PCPPs of interest include two sulfanilamide antibiotics (sulfamethoxazole and trimethoprim), which are exclusively used by humans and are prescribed and dosed together. Sulfamethoxazole has been reported as a common organic contaminant in wastewater (Barnes et al., 2008; Glassmeyer et al., 2005; Kolpin et al., 2002).

In addition to human antibiotics, researchers have detected other pharmaceuticals, such as carbamazepine and acetaminophen (antipyretic/analgesic), in surface water at high frequencies of detection in 82 and 50 percent of samples, respectively (Glassmeyer et al., 2005). The extent to which these compounds reach surface waters from treated or untreated wastewater discharges would be challenging to quantify.

One PPCP that has consistently surfaced in the literature as a viable sewage tracer is carbamazepine because of its chemical characteristics and behavior in the environment. This compound is very stable and soluble in the aquatic environment, is not readily removed during the treatment process, and is specifically derived from human sources (Benotti and Brownawell, 2007; Daneshvar et al., 2012; Doyle et al., 2009; Glassmeyer et al., 2005; Johnson et al., 2012; Nakada et al., 2008).

Carbamazepine is a better candidate than other compounds discussed here because it has an exclusively human source (Glassmeyer et al., 2005). Because of its conservative behavior, researchers have used carbamazepine in groundwater tracer studies to assess the influence of treated wastewater infiltrated through soil (Benotti and Brownawell, 2007; Clara et al., 2004; Daneshvar et al., 2012; Doyle et al., 2009; Johnson et al., 2012; Lubliner et al., 2010). However, because carbamazepine is discharged from wastewater treatment plants unchanged, it is not useful in determining if a source is treated or untreated effluent.

Other human-use pharmaceutical compounds can serve as potential sewage tracers. Metformin, an antidiabetic drug, has one of the highest prescription dosing rates of pharmaceuticals worldwide (grams/day) and has been detected in King County surface waters in concentrations between approximately 200 ng/L and 800 ng/L (King County, 2017). It has also been detected and is considered indicative of the influence of a City of Milwaukee's wastewater treatment plant on Lake Michigan (Blair et al., 2013).

Given the current detections in King County waters along with other research on its modest treatment efficiency and low environmental degradation (Scheurer et al., 2012), it appears that metformin and its metabolites are potentially viable sewage tracers. Locally, however, little is known about metformin concentrations in effluents of concern like CSOs or the number of prescriptions written for the local population. Thus, additional characterization of influent/effluent characteristics and field monitoring of surface waters for metformin would be required before this compound could be used as a reliable sewage tracer.

4.4 Ions and Trace Elements

Researchers have also used specific ions and trace elements as reliable tracers of sewage. Chloride and specific conductance have shown promise on small streams. Direct discharge of sewage can increase both the ionic and chloride levels in freshwater streams (Hyer, 2007). However, chloride and conductivity are unlikely to be useful in larger surface water systems and estuaries. Additionally, chloride and conductivity would not be effective as sewage tracers in the Duwamish Estuary, Lake Union/Ship Canal, or Elliott Bay because of saltwater present in these waterbodies.

5.0 CONCLUSIONS

This literature review identified five compounds that have potential for use as sewage tracers: sucralose, acesulfame, caffeine, carbamazepine, and metformin.

The literature review found that the search for an effective sewage tracer is still ongoing. There is a reliable and inexpensive analytical method for caffeine but not for the other four potential tracers. However, tracing the fate and transport of sewage in surface waters would require using compounds in addition to caffeine. No single compound is suitable for addressing the variety of scientific inquiries for which tracers may be used. The use of several compounds has merit because multiple lines of evidence will improve confidence in the results. A group of compounds exhibiting a variety of chemical characteristics used under different environmental conditions would likely provide a broader range of information than a single tracer. For example, caffeine is removed during the treatment process and so would not be useful for tracing treated wastewater. Caffeine also is present in the environment from sources other than sewage. Tracing caffeine could be useful but a clearer understanding of the fate of sewage in the environment would be improved using chemicals, in addition to caffeine, with different sources and characteristics in the environment.

The most critical element of any future tracer study is to identify specific study questions. Tracer analysis can be expensive and further field investigations into tracers should be guided by focused hypotheses and questions to help ensure that the investment is cost-effective and worthwhile. Advances in analytical chemistry have allowed for the analysis of a variety of new and unique compounds, and several of these show promise as indicators of wastewater influence on receiving waters.

An additional next step would be to identify cost-effective analytical methods for monitoring chemical tracers in the environment. Once identified, these methods could be used to characterize tracer concentrations and their contaminant pathways and to address questions about the fate and transport of sewage in the waterbodies of interest.

Sophisticated statistical software may be useful in evaluating the patterns in a suite of indicator compounds and the diagnostic signature of particular wastewater sources whether untreated or subjected to primary, secondary, or tertiary treatment. One such statistical package is published by EPA as “PMF 5.0” (Norris et al., 2014). Positive matrix factorization (PMF) is used to reduce a large number of variables into unique factors that can then be related to environmental conditions or influences such as treated or untreated CSO discharges.

After a tracer or tracers are chosen and suitable analytical methods identified, collection and analysis of samples from all potential pathways, including stormwater outfalls, treated and untreated CSOs, treatment plant effluent, and upstream inputs, can be done. Characterizing all the possible sources draining to receiving waters is necessary to understand the variety of potential tracer inputs and their relative contributions. Questions about the fate and transport of treated or untreated wastewater could then be addressed.

Examples of focused tracer studies are as follows:

- Providing a line of evidence in investigations of the relative influence of treated and untreated CSOs on receiving waters compared to untreated sewage discharges nearby, such as faulty cross connections with the stormwater system, homeless encampments, or illegal vessel discharges.
- Describing the relative influence of different wastewater or sewage treatment techniques. For example, a smaller treatment plant with minimal treatment technology may have a much larger influence on Puget Sound than a large plant with more robust treatment technology.
- Supporting investigations of failing septic systems and aiding in targeting specific systems for dye testing and then describing the impacts of such failures on streams, rivers, or Puget Sound.
- Monitoring the fate and transport of sewage in surface waters or calculating the relative contribution of raw sewage of surface waters being monitored.

King County will continue to monitor the state of sewage tracer research in the future.

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